Observation of whispering gallery modes in the mid-Infrared with a Quantum Cascade Laser: possible applications to nanoliter chemical sensing

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ABSTRACT

Excitation of the whispering gallery modes (WGM) of a CaF_2 ball resonator is demonstrated at 4.5 micron with a pulsed Quantum Cascade laser. A prism coupling scheme for mid-infrared is described. Future applications of WGM resonators as hyphenated inline chromatography sensors are discussed.

KEY WORD LIST

Mid Infrared CaF2 Whispering Gallery Mode, Quantum Cascade Laser, Gas Chromatography Infrared Sensor

INTRODUCTION

Whispering gallery mode (WGM) resonators have generated lots of interests in the past decade as they can have ultra-high Q factors over a wide wavelength range in a tiny cavity, which usually could not be realized with conventional cavities with dielectric coatings[1, 2]. The paths of the WGMs lie very close to the surface of the resonator with a portion of the electromagnetic (EM) wave traveling outside, i.e. evanescent wave. Losses due to absorption or scattering of the evanescent wave outside the resonator will change the Q factor of the resonator. Several groups demonstrated that WGM resonators could be used for absorption sensing in gases and liquids, detection of refractive index changes around the resonator and single molecules on its surface[3, 4]. All chemical sensing studies performed until now with WGM used near-IR and visible continuous wave (CW) single frequency lasers because of the wide availability of such lasers and detectors. However, mid-Infrared (MIR) and far-Infrared (FIR) regions of the spectrum are much more important for chemical sensing because the fundamental bands of vibrations lying in these regions are 1~3 orders of magnitude stronger than their near-infrared overtones. However, there are some difficulties in realizing MIR/FIR coupled WGMs., i.e. there are very few choice of single mode fibers in the MIR/FIR, and the most common fused silica material is not transparent in that range, and finally CW QCL sources are still far from as popular as telecom CW diode lasers, but the latest improvements in QCLs bode well for their applications.

Because of the high brightness of QCLs, we recently demonstrated that the radiation of a QC laser could be efficiently coupled into a 300μm diameter hollow waveguide and used as a microliter gas sensor in gas chromatography. Here, we demonstrate that pulsed QCLs, in spite of their relatively large linewidth , could be efficiently coupled into WGM cavities. We demonstrate, to the best of our knowledge, the first excitation of a WGM resonator in the MIR and discuss conducting cavity enhanced spectroscopy measurement in the IR with such WGMs.

EXPERIMENT

The schematic of the setup is given in Figure 1. A Quantum Cascade (QC) Distributed Feedback (DFB) pulsed laser operating at 4.45micron (tunable from 2,259cm^{-1}~2,264cm^{-1}, Alps Lasers, Switzerland) is used in this experiment. The laser is operated at 800 kHz and has a peak power about 100mW, and pulse width is about 25nsec. The manufacturer reported linewidth is about 0.01cm^{-1}. The laser output is focused with a simple bi-convex ZnSe lens at the hypotenuse face of a ZnSe right angle prism about 4" away from the laser housing. A 5mm diameter CaF_2 ball lens (part #: CF-B-5 from ISP Optics, Irvington , NY) is mounted on a precise 3-axis translation stage fitted with

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actuators with resolutions of 0.05μm/step. The ball lens is positioned carefully to approach the Total Internal Reflection (TIR) spot of the laser beam on the hypotenuse face of the right angle prism. The laser beam experiences two TIRs in the prism, before exiting it and propagating toward a MCT detector. The MCT detector is a thermoelectrically cooled version made by VIGO (Model: PDI-2TE-5, with peak detectivity, D*, of 7×10^{10} cmHz$^{1/2}$/W at 4.5μm when operated at -30°C, Vigo Systems, Warszawa, Poland) and fit with a preamplifier (model 480, by Boston Electronic, Brookline, MA, USA). The signals from the detector are sampled at the peak of the laser pulse by a Sample&Hold circuit, custom built upon OPA615 (Texas Instrument) chip. The signal is then converted to digital format by a data acquisition card (Model 6011E by National Instrument, TX). The QCL wavelength is scanned by a ramp generator, which modulates the bias current to the QCL, thus changing its wavelength. The ramp function is a triangle waveform with a short window at the end of each ramp period which turns off the current pulses to measure the detector baseline.

We carefully adjusted the angle of incidence (AOI) of the QCL beam at the hypotenuse face so that only the lowest order WG modes, i.e. $q = 1$, are excited. We achieved near single mode excitation for $q = 1$ WG modes when we adjust the AOI of QCL at the hypotenuse to be slightly above the critical angle for total internal reflection to happen at the ZnSe and CaF_2 interface, which is 35.3° at 4.5μm wavelength. As AOI getting closer to this critical angle, the WGM structure starts to simplify. In our case, we start to see only one major dip remains strong in one Free Spectra Range (FSR) of the ball cavity; and at AOI larger than this critical angle, only one major dip stays, and itself also getting weaker very fast as the AOI gets above the critical angle.

Following the established procedures for fabricating high Q CaF_2 WGMs[5], we also fabricated CaF_2 and BaF_2 discs with different morphology on the outskirt. Figure 4 and 5 shows the SEM images and also the Qs demonstrated for the 5mm ball lens, and home fabricated 5mm discs.

RESULTS

Figure 2 shows the WGM mode dips getting stronger as the position of the ball lens getting closer to the hypotenuse face. A maximum WGM coupling induced loss of 20% is observed when optimized. The spacing between the major WGM mode dips is measured to be ~0.45cm^{-1}, which is calibrated with an air spaced Ge etalon with a Free Spectral Range of 0.075cm^{-1}. This value is consistent with the size of the CaF_2 ball lens, i.e. 5mm diameter. In fact, we noticed that the WGM modes for this 5mm CaF2 ball are much simpler at 4.46μm, when compared to the case at 1.55μm, i.e. with the same setup at the critical AOI for 1.55μm as shown in the insert of figure 2. We attribute this reduction in the number of WGM dips to the facts that we are using a 4.46μm laser, instead of a 1.5μm laser. The longer wavelength laser reduces the equator's WGMs to almost single q=1 operation under the critical AOI coupling. The reduction in the number of WGM modes is also a result that we are using TE polarization only because the input is from a TM polarized QCL laser chip, whereas the input is usually not linearly polarized from the fiber coupled 1.5μm telecom laser to the CaF_2 spheres.

We also notices that the WGM dips' linewidth at 4.45μm goes up from ~0.1cm-1 to ~0.2cm-1 as we move the ball lens closer to the prism surface. This broadening is explained as a result of overloading as we explained below. At 2,240cm^{-1}, or 4.46μm, using the width of the dips in the transmittance spectrum, i.e. ~0.01cm^{-1} when under coupled, we estimate the Q factor to be approximately 2×10^5. However, this Q value must be underestimated because we are using a pulsed laser and the inherent line width is quite wide already, i.e.0.01cm^{-1} or 300MHz, and the Q therefore will have an upper measurable limit of 2.2×10^5. We also observed that the line width of the WGM dips broadens as we made the QC laser pulse longer, which introduced larger frequency chirp. As verification to the exact Q value, we also measure the linewidth of the WGM dips at 1.55μm under critical AOI conditions, and get a linewidth about 0.02cm^{-1}, and this translates into a Q of 3×10^5 at 1.55μm, see the top left insert in figure 2.

The low Q value at 1.55μm is attributed to the rough surface quality of the ball lens, which is specified as 60-40 scratch and dig, although usually much better. The microscope view of the ball confirms that majority of visible surface digs are ~20μm in cross sections, see bottom right insert in figure 2. Since the energy loss in the ball is dominated by scattering loss, and Q is therefore proportional to the cube of the wavelength, as given in equation[6]:

$$Q \approx \frac{3\lambda^3 a}{8n\pi^2 B^2 \sigma^2}$$

where λ is the wavelength, a is the radius of the WGM cavity, n is the refraction index of the resonator material, B is the correlation length, and σ is the roughness. Here, λ = 1.55 μm, a = 2,500 μm, n = 1.426. If we use B = 200μm, and σ =5 μm for the 20-10 scratch and dig quality, Q would be only 0.00025. In order to justify the 3×10^5 value we observed, we have to use B * σ =0.028 μm^2. This adjustment might be reasonable because the WGMs propagate along a small portion of the surface of the ball. For this set of parameters, our Q should be close to 10^7 at 4.55μm. With such high Q value at 4.55μm, our pulsed QCL will have a very low coupling efficiency, and this explains that we see very little dips when we start to observe WGM features, although the small dip depth might also be a result of the fact that we are still far away from the prism and coupling is really poor. As we get closer, or even touching the prism surface, overloading happened and causes the Q to drop and therefore increased coupling efficiency and larger dip depth. To fully utilize and verify the high Q value available at 4.46μm, a CW QCL should be used in future experiments. If the Q is indeed as high as 10^7 at 4.46μm with such a low quality CaF_2 ball lens, then this means WGM sensors could be very tolerant to dust or impurities in the bulk environment.

DISCUSSIONS

Sensitivity

As demonstrated with fluorite crystalline WGM cavities in the near IR, a CaF_2 WGM resonator could potentially achieve values over 10^9 for the Q factor at 1.5μm after careful polishing and annealing. Because of the longer wavelengths in the MIR and FIR, the main Q limiting factor will no longer be the surface scattering loss but the material absorption. This lower requirement on surface quality will make WGM sensors more robust in the MIR/FIR, i.e. they would be less sensitive to dust and environmental changes. CaF_2 already starts to have higher absorption at the wavelengths over 4μm and limit its Q at 4.5μm to below 10^9, while BaF_2 could be a better candidate if we need to use longer IR wavelengths or get even higher Q factors at 4.5μm.

Although fluorite crystalline WGMs resonators potentially have very high Qs in the MIR/FIR, this does not translate into very long effective path length for chemical sensing, especially if we try to sense gas phase chemicals. This is because the evanescent wave only accounts for a small portion of the Electro Magnetic (EM) field of the WGMs, and only the evanescent wave interacts with external chemicals. The fraction of the WGM's energy in the evanescent field depends on the refractive indices inside and outside resonator. For gas phase sensing, this fraction, f, is only ~1% or less[4]; while for liquid phase, f could be as high as 30%[3]. Therefore, with the Q value of $>10^8$ in the gas phase and $>10^7$ in the liquid phase, the effective sensor path length would exceed 1 meter. One meter path length would be sufficient for liquid sensing but in the gas phase it is much shorter of the path lengths provided by multipass cells (1~>100 meters) and cavity enhanced methods (>1,000meters). However, WGM sensors could be used in applications that require small sample volume or overall small size of the sensors. For example, they could be used as inline sensor for Gas Chromatography (GC). With a one meter path length one could detect ppbV concentrations of CO_2, which would exceed the sensitivity of the mainstream GC sensors[7].

The sensitivity of WGM sensors could be significantly improved by engineering WGM resonators that have a large portion of the mode energy outside the cavity . This was demonstrated with a subwavelength slot waveguide for TE polarized light [8]. The detection of external gas refractive index change was enhanced over 20 times because 80% of the mode energy was concentrated in the slot. The advantages of using slot waveguide WGM resonator in the MIR and FIR are obvious. First, the slot width could be as wide as a micron or larger which allows fast exchange of samples through diffusion processes, and the larger dimensions also make fabrication tolerances less strict. Second, MIR and FIR wavelengths are also much more tolerant to rough surface finishes when fabrication involves etching, which has been the major limit in achieving high Qs in WGM cavities fabricated with mask and etching. For the quality factors $>10^8$ in the MIR and FIR, which could be realistically achieved for several fluorite crystals and also ZnS or ZnSe materials, and the f factor to over 50% (for a slot WGMs resonator), one could have an effective path length well over 10 meters.

Measurement technique

Building WGM resonator based sensors is not straightforward because the WGM resonances must be tuned to the absorption features of detected species and/or the laser wavelength. A locking and scanning scheme has already been demonstrated in the Near-IR gas sensing experiment[4, 9].

A pulsed QCL laser is not suitable for this discrete frequency locked measurement because linewidth of a pulsed laser is usually too wide to see such dips when the Q is really good, e.g. over 10^7. So, a Continuous Wave (CW) QCL with single frequency output is required.

Another interesting analogy to the latest traditional cavity enhanced spectroscopy, i.e. Off-Axis Integrated Cavity Output Spectroscopy (OA-ICOS)[10], has been demonstrated with WGM cavities[11]. By engineering the WG cavity so that the mode distribution is essentially continuous for external tunable laser while maintaining the high Q values ($>10^8$) for this continuous WMG. In this way, it is very simple to do tunable laser spectroscopy in the IR with WGMs, both pulsed and CW QCLs could be used and no frequency locking scheme is needed.

In both schemes, when the external loss due to gases is small and WGM is under coupled, the dip depth will get smaller when there is external loss due to gas absorption, i.e. we will see the transmission spectrum showing peaks due to gas absorptions.

Sample volume and integration with chromatography instrument

One of well known advantages of WGMs is that they require minimal sample volume; it can be as low as a nano-liter. This compares favorably with other well established cavity enhanced or multi-pass absorption instruments which need over 100mls of samples. Gas chromatography could be one the application areas for WGM sensors. GC columns are usually only a few hundreds of microns in diameter. The column diameter can be increased locally but this would require the use of a makeup gas to preserve the gas peak width and lead to sample dilution. The gas peaks in the GC column can be separated by only a few centimeters. This puts a limit of the length of the hollow waveguide that can be used in the sensor. The WGM sensors could achieve the path length of over a meter any compromise in the chromatography temporal resolution. Figure 3 shows some of the deployment schemes of such WGM cavities with inline capillary, where the dead volume is minimized while coupling the interaction of the WGM cavity with the carrier gas/fluid and tunable lasers.

We gratefully acknowledge the helpful discussions on WGMs with Dr. Ivan Grudinin, and on slot waveguide and optofluidic structures with Dr. Zhenyu Li. Further sensing research using broadband WGMs based on fluorite crystals are being carried out at our center.

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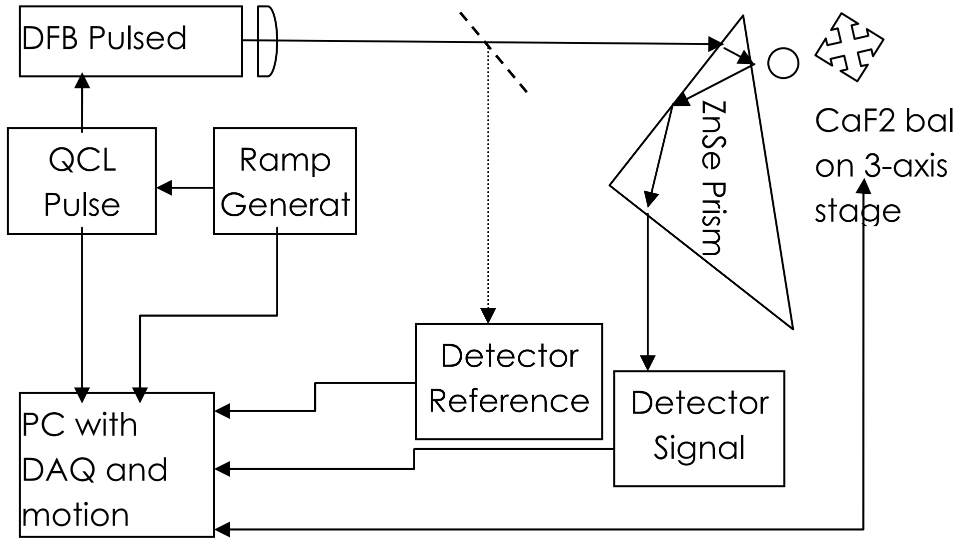


Figure 1. Schematic of the QCL coupling to WGM setup. The ZnSe prism is used at critical TIR angle to favor the evanescent wave coupling into the lowest WGM modes of the CaF2 ball lens' cavity.

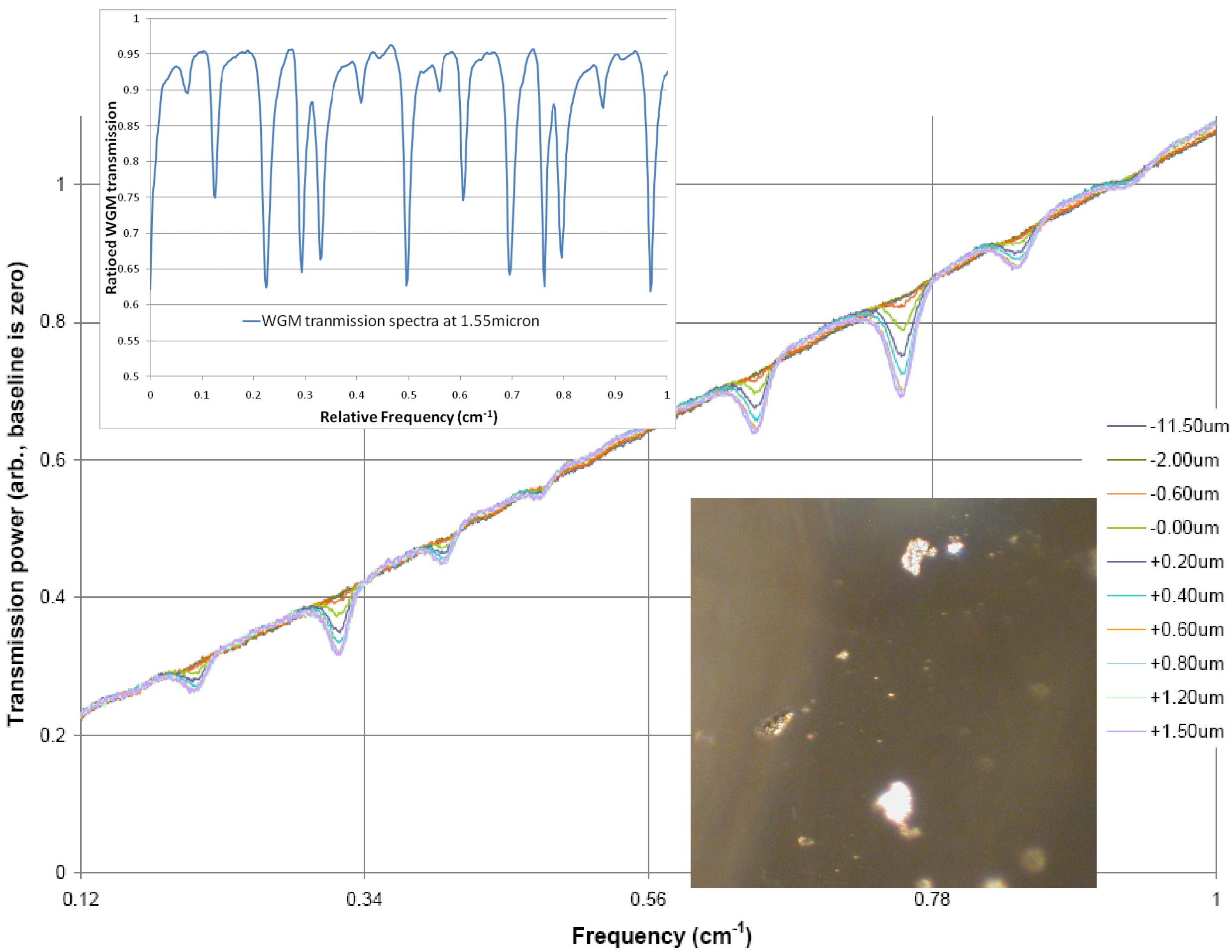


Figure 2. Transmitted QCL laser power spectra v.s. the distance of the CaF2 ball lens to the hypotenuse face of the ZnSe Right Angle Prism. The top left insert is the transmission spectra of a tunable 1.55µm laser, and the bottom right insert is the microscope view of the surface quality of the CaF2 ball lens, the size of the view is 60x60µm.

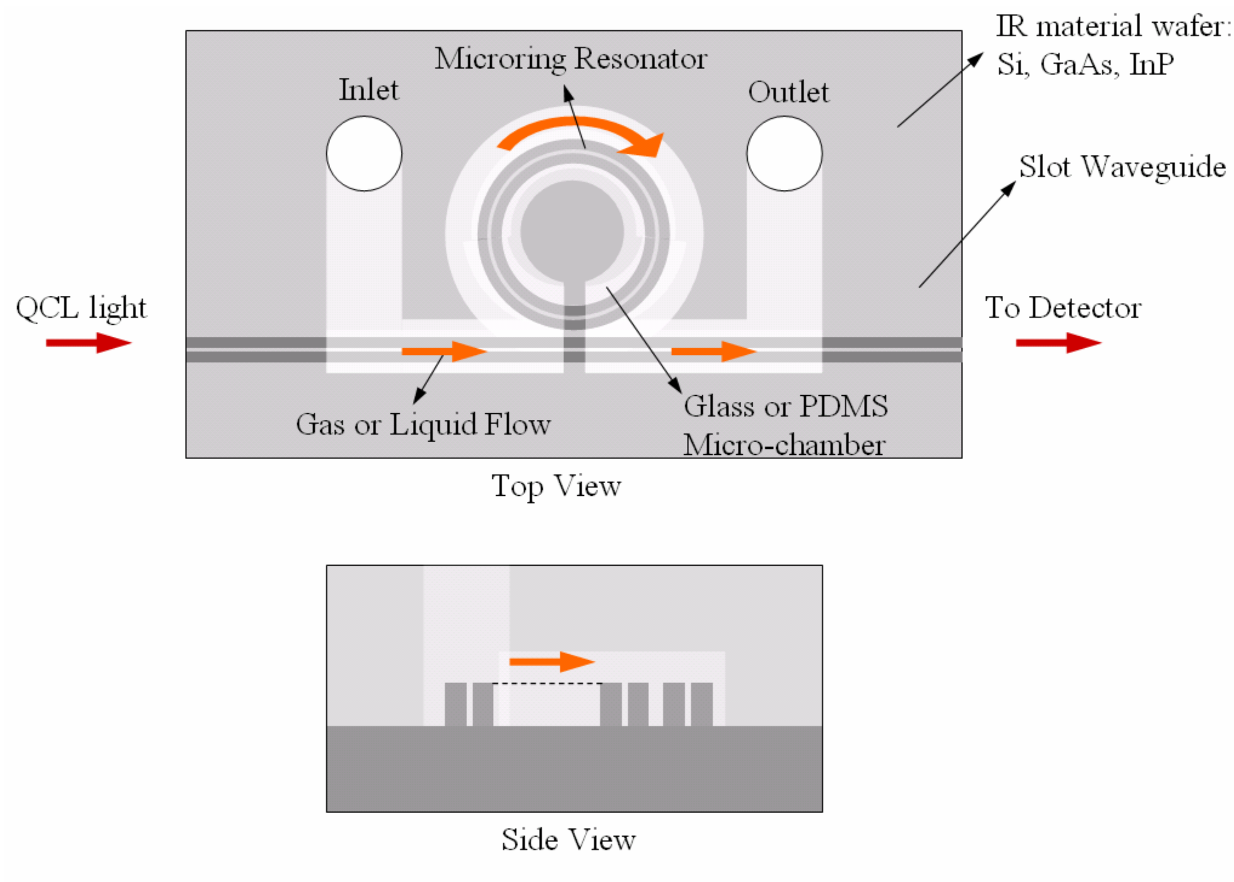


Figure 3. Proposed scheme for coupling QCL laser into ring shape WGM cavity (a CaF2 disc or a microfabricated ring slotted waveguide) and also flow chromatography capillary around the cavity. The distance between the IR laser beam waveguide coupler and the ball could be less than 1µm and this will serve as an effective flow stopper and enable uni-directional fluidic flow around the round cavity. The total length of flow around the round cavity is about 10mm, and cross section is ~0.100mm^2, and the volume is therefore about 1µl.

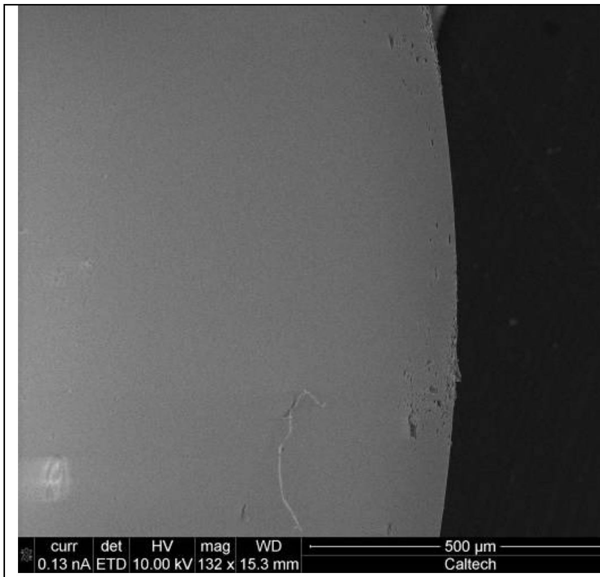


Figure 4A: Commercial 5mm ball lens morphology that support the WGM modes (SEM)

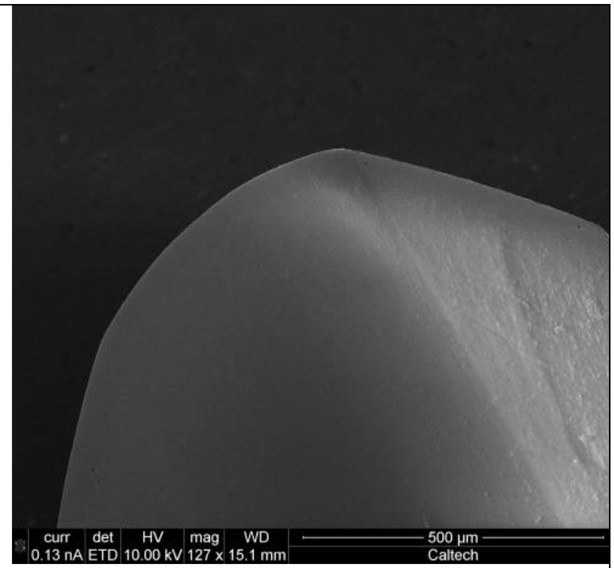


Figure 5A: 5mm diameter 1mm thick home polished disc morphology that support the WGM modes (SEM)

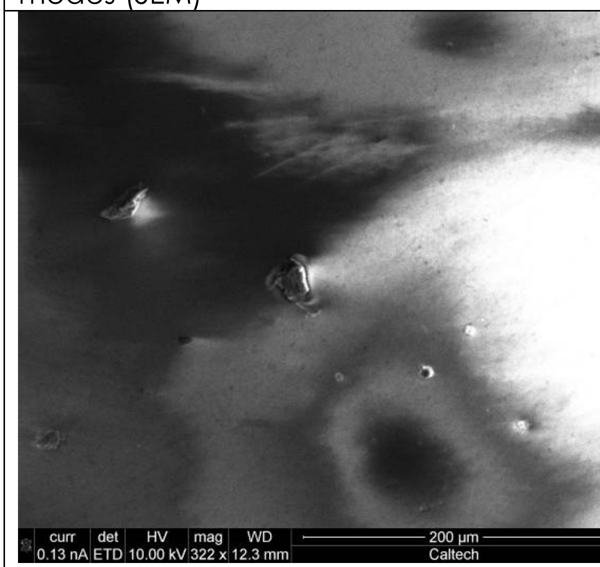


Figure 4B: 5mm ball lens surface quality has large dents at over 20µm in size

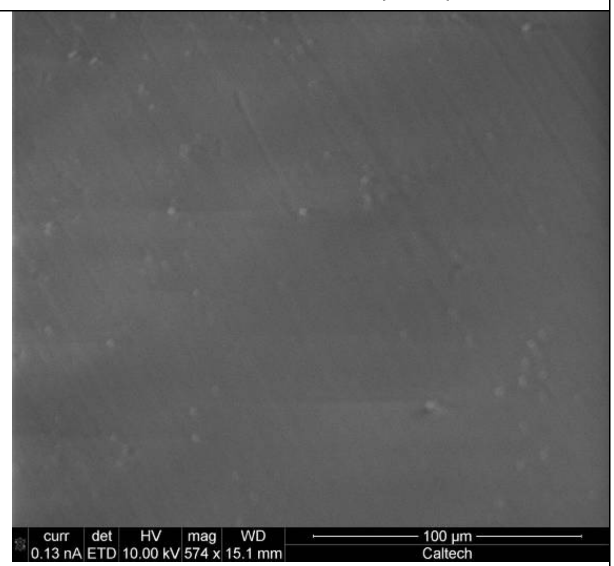


Figure 5B: 5mm diameter 1mm thick surface quality has long thin and low level scratches at over 20µm in length

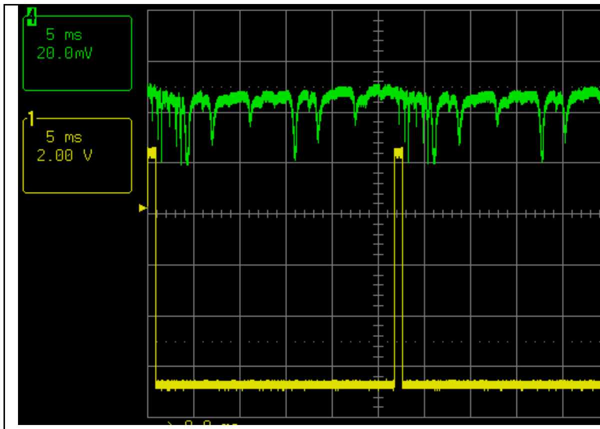


Figure 4C: 5mm ball lens, FSR is 0.47cm-1 or 11.5msec, and the FWHM of the dip is 0.2msec, see figure below

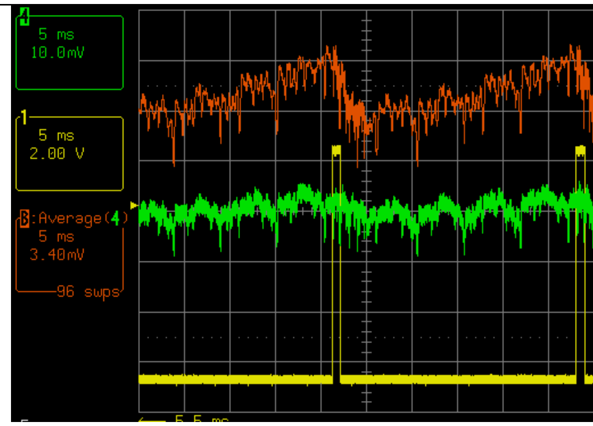
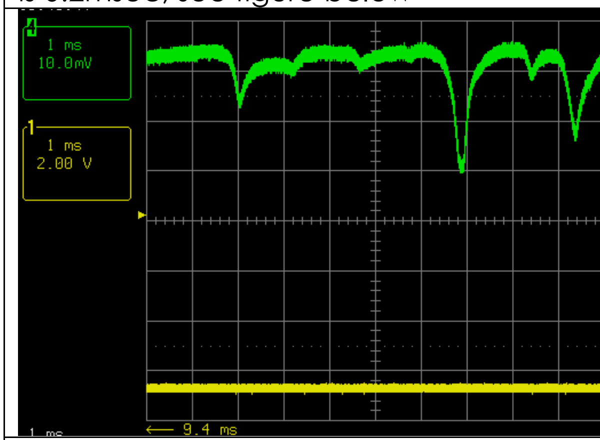
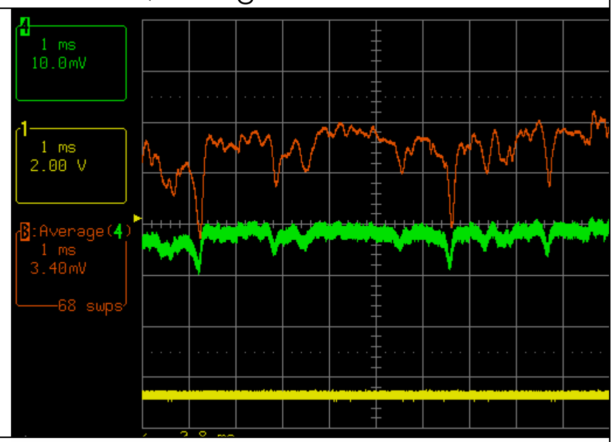


Figure 5C: 5mm dia. 1mm thick home polished disc, and the FWHM of the dip is <0.1msec, see figure below





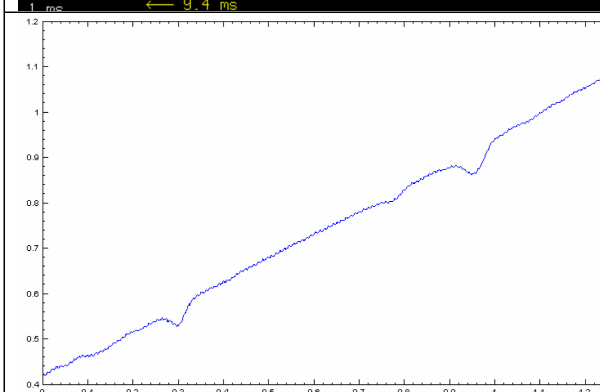


Figure 4D.The 5mm ball lens WGM spectra excited with QCL at 4.5μm

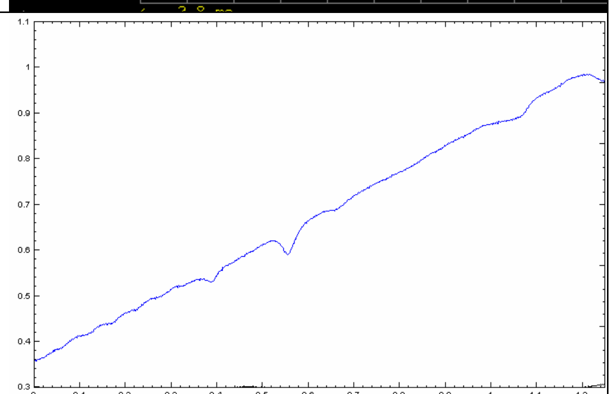


Figure 5D. The thin disc WGM spectra excited with QCL at 4.5μm